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Dr. Moeljanto Leksono and Dr. Jacques I. Pankove

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Astralux, Inc.
2386 Vassar Drive
Boulder, CO 80303

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The first step in growing a GaN boule was to demonstrate the possibility of growing GaN from elemental Ga and ammonia on a substrate. Although GaN crystals were obtai the effusion rate of Ga vapor was too high. A number of other problems was uncover and their solution was conceived. Several changes to the deposition system were made, but to succeed, a more radical redesign is needed. This new design will be proposed as a first task during Phase II. The Phase I work has been a very useful experience that uncovered valuable insights.

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Growth of GaN Single-Crystal Boules

Contract Number: F49620-94-C-0067
Final Report from Astralux, Inc., Boulder, Colorado

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Abstract

The first step in growing a GaN boule was to demonstrate the possibility of growing GaN from elemental Ga and ammonia on a substrate. Although GaN crystals were obtained, the effusion rate of Ga vapor was too high. A number of other problems was uncovered and their solution was conceived. Several changes to the deposition system were made, but to succeed, a more radical redesign is needed. This new design will be proposed as a first task during Phase II. The Phase I work has been a very useful experience that uncovered valuable insights.

This project was sponsored by BMDO/IST and monitored by AFOSR/NE.

Introduction

The motivation for this project is the need for high quality GaN. Everyone working with this material recognizes the fact that GaN grown on sapphire has an enormous density of defects, because of a 16% lattice mismatch. These defects lower the carrier mobility, introduce non-radiative recombination centers, and generate unwanted carriers in the presence of an electric field. These defects are detrimental to many applications such as injection lasers, UV detectors, and high temperature electronic devices.

A 3% lattice mismatch is obtained when silicon carbide, SiC, is used as the substrate. Unfortunately, SiC is extremely expensive and is still plagued by the presence of micropipe defects. To avoid these problems one needs to resort to homoepitaxy and grow GaN on GaN. Our purpose is to provide the starting GaN substrate. For this, a simple approach had been envisioned that could lead to the continuous growth of a single crystal. Later, this single crystal or boule could be sliced for subsequent homoepitaxy.

New Approach

The present approach is to start with a vapor of elemental gallium, Ga, that is reacted with nitrogen from ammonia, NH_3. For this, several versions of the reactor originally proposed for Phase I were tried (Fig. 1). Ga was to react with NH_3 to form GaN on the sapphire substrate. If this were successful it could lead to a faster growth rate and a purer material than possible with MOCVD. Of course, the material would still contain stress-induced defects. However, the stress content would decrease as the GaN grew thicker, and one could use GaN as the next seed instead of sapphire.

Experience with the first system uncovered many unforeseen problems that needed to be solved one by one at great cost in time and money. In retrospect six months to accomplish the Phase I goal was not realistic, but the prospect for success is still excellent and the potential benefits make growing single crystal GaN worthy of further effort.

The Challenges

Choice of Ceramic Crucible: Boron nitride, BN, is inert, stable and readily available. But it is fragile and has a high heat conductivity. Fragility is compensated for by extra care in handling. The high thermal conductivity meant that the substrate heater caused faster evaporation of Ga than reaction with NH_3, resulting in a leather-like flexible thick Ga layer containing microcrystals of GaN. Although these crystallites had strong narrow bandedge luminescence (Fig. 2) it was not the desired large area crystal. The thermal coupling between the substrate heater and the Ga reservoir was reduced by machining an external groove between the Ga crucible and the substrate holder. (See Fig. 3.)

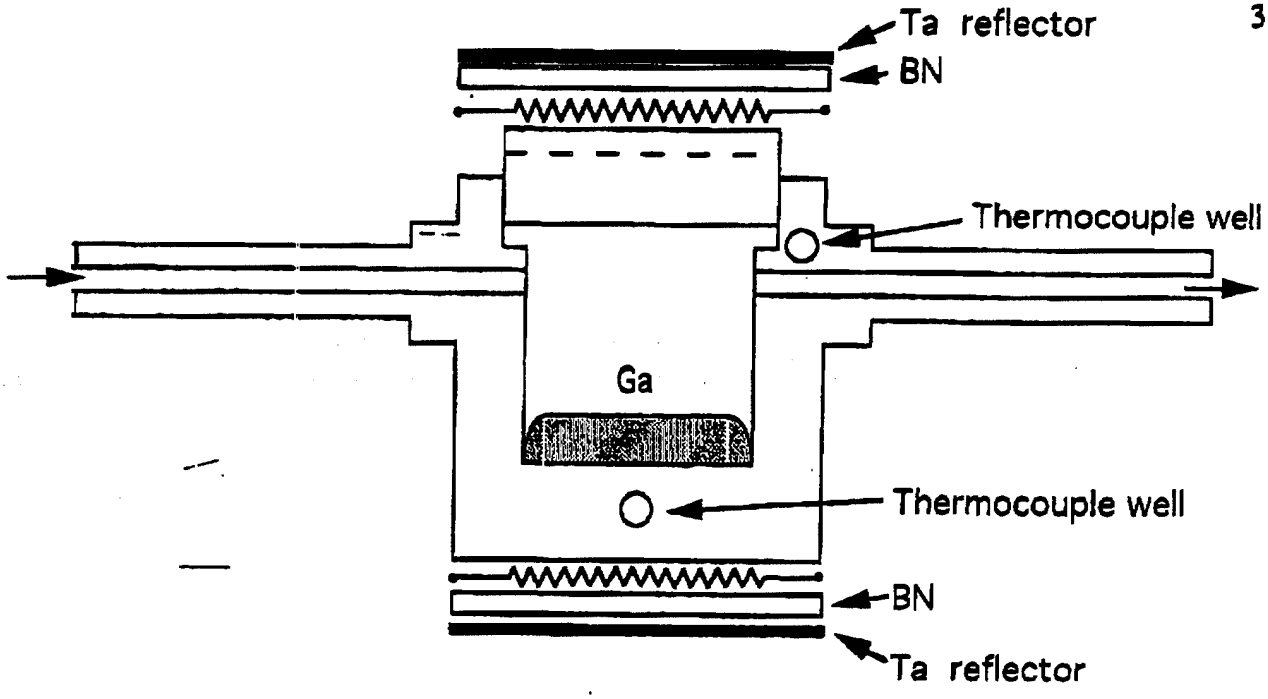

Fig. 1 Schematic Diagram of Initially Proposed Deposition System.

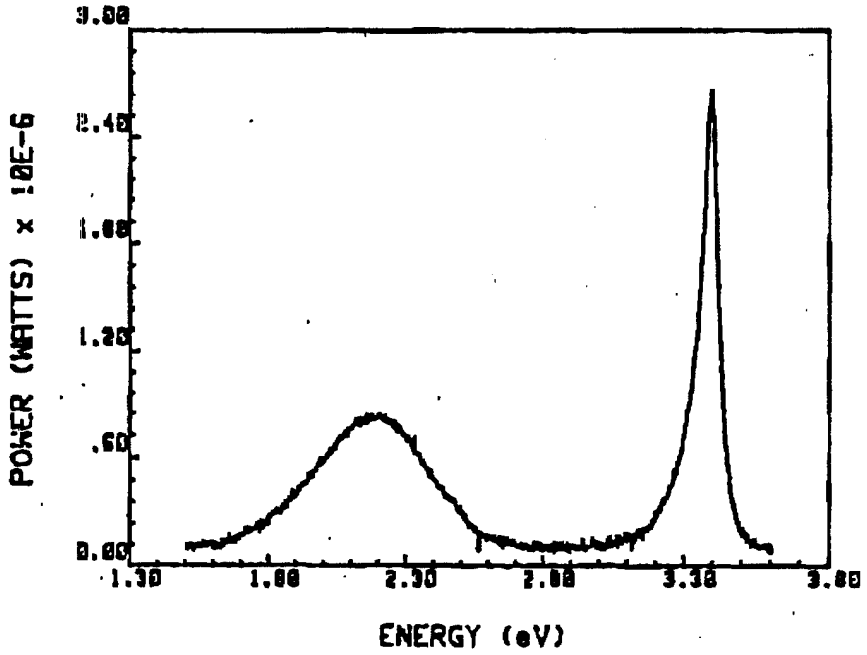

Fig. 2 Room Temperature Cathodoluminescence Spectrum of a GaN Layer Obtained in the Boule Growth System

Choice of Heating Elements: Kantal wire was the first material to be used to form the heating element. It alloyed with Ga and fell apart. Molybdenum was tried next. It too reacts to Ga vapor, but more slowly. The heater was encased in the BN and openings were sealed with a BN cement. Tungsten was tried but it is too brittle for bending and difficult to spot weld. Hence, Mo was chosen for the heating elements.

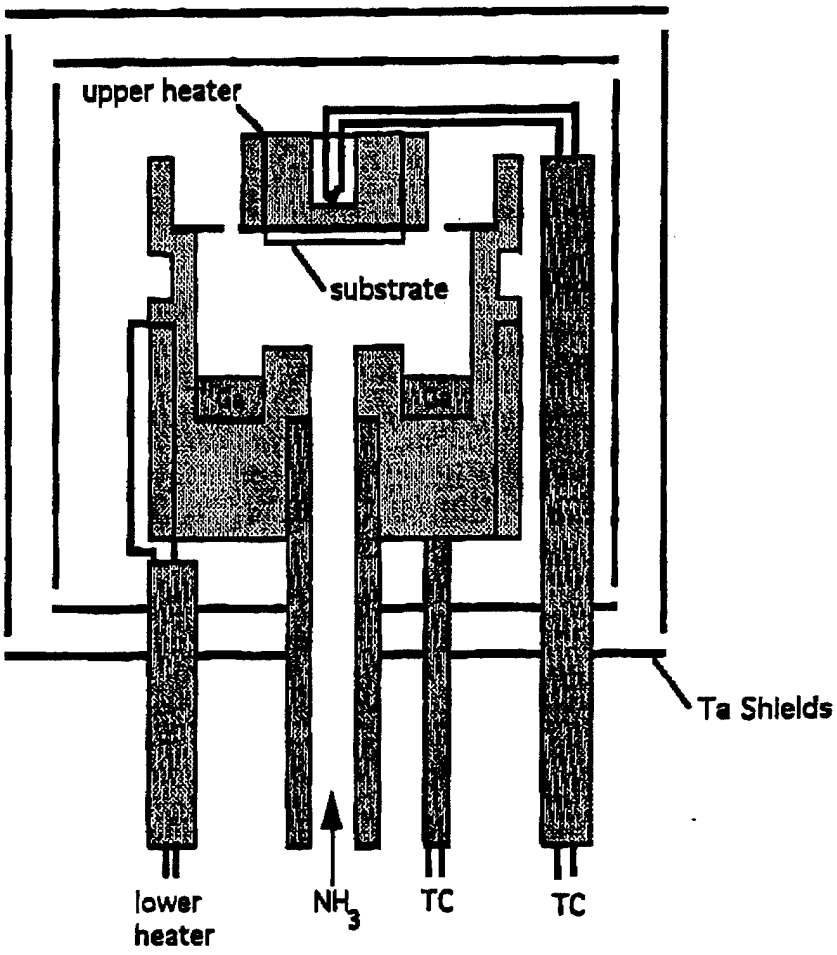

Fig. 3 Schematic Diagram of Second Deposition System.

NH_3 Reactivity: Ammonia dissociates at high temperature (~ 900 C). The proximity of the Ga pool results in too high a Ga vapor pressure. Then the hot filament method was resorted to for decomposing NH_3 near the substrate. An Mo filament reacted with the Ga vapor; W was too difficult to spot weld to the biasing leads. Rhenium, a favorite catalyst,[1] was tried and appeared promising.

Since the heater feed-throughs comprised only four leads, another challenge was how to install three heating elements. Two pairs of leads were connected to the secondary windings of transformers and used to bias the top and bottom heaters. The hot filament was connected between a lead from each heater and a third transformer was inserted between the two heater transformers (Fig. 4).

1 S.N. Foner and R.L. Hudson, J. Chem. Phys. 80, 4013 (1984).

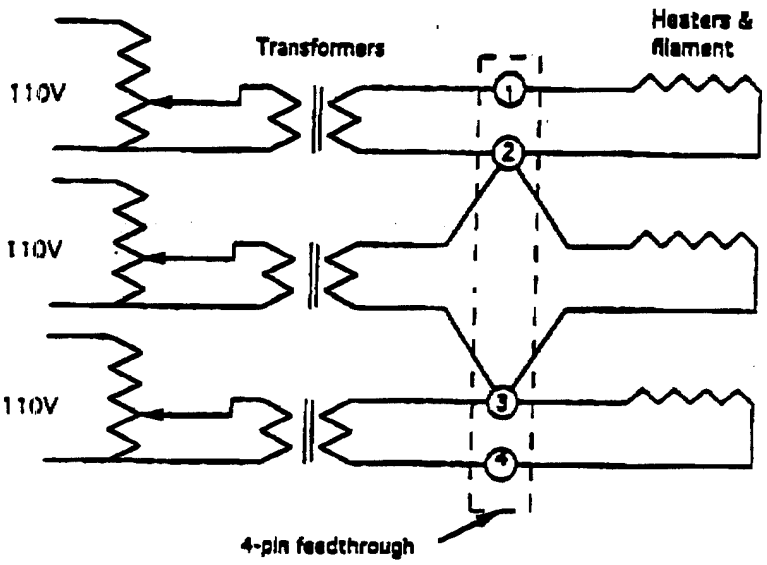

Fig. 4 Wiring Diagram for Driving Three Heaters through a Four-Lead Feed-Through.

With three separate heat sources capable of interaction, the control of each temperature becomes more complex. This challenge will be addressed in an already designed new system to be built during Phase II.

Substrate: Sapphire was used as a substrate for preliminary runs because of convenience. SiC is a better substrate, and tungsten or molybdenum may be even better. Tungsten has a lattice mismatch of only 0.6% and Mo 1.3%. We used a Mo substrate holder and obtained small brick-shaped GaN crystals (Fig. 5). Mo has a cubic structure that nearly matches cubic GaN. A number of randomly arranged parallelepipeds were obtained, as would expected from a poly moly substrate. The GaN parallelepipeds were too small to check by x-ray diffractmetry. Single crystal tungsten is available, costly and requires a 2-month delivery. An order has already been placed for a quicker start if a Phase II contract is obtained. For some applications cubic GaN would be preferable to wurtzitic GaN, for example for edge-emitting lasers where facet cleavage is desirable.

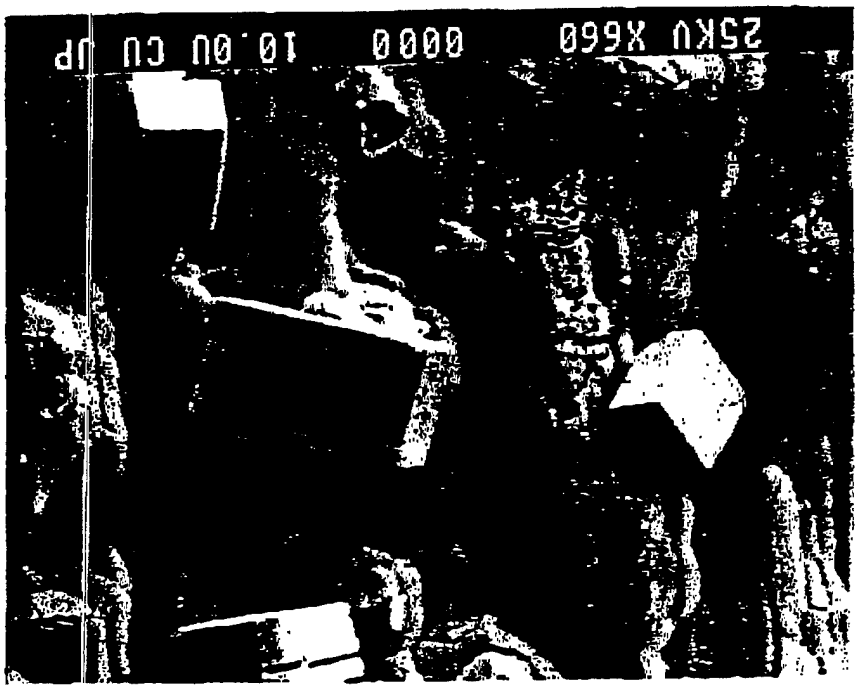

Fig. 5 GaN, posited cubic, grown on Mo.

Phase II Projections

For a Phase II continuation of this project a multiple task approach will be proposed with a timeline that will permit evaluation of performance and assessment of probability of success in the next task.

Task 1: Build and test a new three-zone systems (month 8)

Task 2: Scale up the size of grown crystal (month 13)

Task 3: Build continuous growth system (month 22)

Task 4: GaN slice availability (month 24)

Related Effort

Vodakov, et al. used GaN powder and NH_3 in a thermal gradient to transport Ga from the powder to a closely spaced substrate. A growth rate of 70 μm/hr was obtained.[2] We heard that a group in Berkeley is also transporting Ga from a liquid to a substrate with an NH_3 carrier.

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2 Yu. A. Vodakov, M.I. Karklina, E.M. Mokhov and A.D. Roenkov, "Growth of GaN Epitaxial Layers on Sapphire and SiC Substrates," Izv. Akad. Nauk SSSR, Neorg. Mater. 16 (3), 537 (1980).

Conclusion

Because of unexpected problems, it was not possible within six months to succed in producing single crystal GaN wafers that could be used for homoepitaxial growth of GaN. But this work was very valuable to define the problems and to consider their solutions. It is likely that the successful completion of this new high potential technology may require about eight more months of work. This will be proposed for incorporation as the first task in a Phase II effort.